

High-accuracy calculation of black-body radiation shift in ^{133}Cs primary frequency standard

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Black-body radiation (BBR) shift is an important systematic correction for the atomic frequency standards realizing the SI unit of time. Presently, there is a controversy over the value of the BBR shift for the primary ^{133}Cs standard. At room temperatures the values from various groups differ at 3×10^{-15} level, while the modern clocks are aiming at 10^{-16} accuracies. We carry out high-precision relativistic many-body calculations of the BBR shift. For the BBR coefficient β at $T = 300\text{K}$ we obtain $\beta = -(1.708 \pm 0.006) \times 10^{-14}$, implying 6×10^{-17} fractional uncertainty. While in accord with the most accurate measurement, our 0.35%-accurate value is in a substantial, 10%, disagreement with recent semi-empirical calculations. We identify an oversight in those calculations.

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Since 1967, the SI unit of time, the second, is defined as a duration of a certain number of periods of radiation corresponding to the transition between two hyperfine levels ($F = 4$ and $F = 3$) of the ground state of the ^{133}Cs atom. In 1997, this definition has been amended, to specify that the above statement refers to the atom at a temperature of 0 K (and at rest) [1]. The atomic clocks are usually operated at room temperatures and the specific reference to $T = 0$ K implies that a proper correction for the action of the thermal bath of photons on the atomic energy levels is explicitly introduced. For ^{133}Cs clocks, it is an important systematic correction [2], as the resulting fractional correction to atomic frequency due to black-body radiation (BBR) at $T = 300$ is in the order of 10^{-14} . Moreover, presently, there is a controversy over the value of the BBR shift for the primary ^{133}Cs standard. At $T = 300$ K the values from various groups [3, 4, 5, 6, 7] differ at 3×10^{-15} level, while modern Cs clocks aim at 10^{-16} accuracies [8].

The persistent discrepancies in the BBR shift have prompted the efforts at the US National Institute for Standards and Technology (NIST) on removing the sensitivity to BBR by operating the primary standard at cryogenic temperatures [9] (BBR shift scales as T^4). However, because of the weight limitations, this direct approach would be hardly feasible if next-generation atomic clock were to be operated at the International Space Station [8]. This ongoing controversy and implications for atomic time-keeping serve as motivations for our paper. Here we compute the ^{133}Cs BBR shift using high-accuracy relativistic many-body techniques of atomic structure. Our evaluated error in the BBR shift implies 6×10^{-17} fractional uncertainty in the clock frequency with the value of the BBR shift consistent with the most accurate (0.2%-accurate) measurement [3]. However, our 0.35%-accurate value is in a substantial 10% disagreement with recent semi-empirical calculations [6, 7]. We show that this discrepancy is due to contributions of the intermediate continuum states omitted in those calculations.

First, let us review underlying theory of the Cs BBR

shift. BBR causes a weak oscillating perturbation of atomic energy levels. Conventionally, the leading term in the BBR contribution is parameterized as the fractional correction to the unperturbed clock frequency, $\nu_0 = 9\,192\,631\,770$ Hz,

$$\delta\nu^{\text{BBR}}/\nu_0 = \beta \times (T/T_0)^4,$$

where $T_0 = 300$ K. Evaluation of the coefficient β is the goal of this work. This coefficient can be related to the scalar differential polarizability for the hyperfine manifold of the $6s_{1/2}$ Cs ground state. Indeed, the characteristic thermal photon energy at room temperatures is much smaller than the atomic energies, so that the perturbation can be well described in the static limit. Moreover, contributions of electro-magnetic BBR multipoles beyond electric dipoles, as well as retardation corrections, are highly suppressed [10]. Then the BBR shift of the energy level is given by (atomic units are used throughout, α is the fine-structure constant)

$$\delta E_F^{\text{BBR}} \approx -\frac{2}{15}(\alpha\pi)^3 T^4 \alpha_F(0), \quad (1)$$

where $\alpha_F(0)$ is the static scalar electric-dipole polarizability of the hyperfine level F . The vector and tensor parts of the polarizability average out due to the isotropic nature of the BBR.

The relation of the BBR shift to polarizability has been exploited in the most accurate measurement of the differential Stark shift [3]. However, recent direct temperature-dependent measurement [5] of the BBR shift turned out to be differing by about two standard deviations from the indirect measurement. Namely this difference has stimulated the recent interest in the Cs BBR shift.

The overall BBR shift of the clock frequency is the difference of the individual shifts for the two hyperfine states ($F = 4$ and $F = 3$) involved in the transition. While taking the difference, the traditional lowest-order polarizability of the $6s_{1/2}$ level cancels out and one needs to evaluate the third-order F -dependent polarizability

$\alpha_F^{(3)}(0)$. This contribution involves two E1 interactions $V = -\mathbf{D} \cdot \mathbf{E}$ with the external electric field and one hyperfine H_{hfs} coupling [11]. We parameterize the general correction to the energy as a sum of four diagrams,

$$\delta E_F^{(3)} = \langle F | H_{\text{hfs}} R V R V | F \rangle + \langle F | V R H_{\text{hfs}} R V | F \rangle + \langle F | V R V R H_{\text{hfs}} | F \rangle - \langle F | H_{\text{hfs}} | F \rangle \langle F | V R R V | F \rangle,$$

where $R = (H - E_v)^{-1}$ is the resolvent operator, with H and E_v being the atomic Hamiltonian and the ground state energy, respectively. The four contributions will give rise to top, center, bottom, and residual contributions discussed below. (Naming convention reflects relative position of the hyperfine operator in the string of the three interactions). The top and bottom diagrams are equal due to hermiticity considerations. Due to angular selection rules only magnetic dipolar interaction remains; we write $H_{\text{hfs}} = \boldsymbol{\mu} \cdot \mathcal{T}^{(1)}$, where μ is the nuclear magnetic moment and $\mathcal{T}^{(1)} = -i\sqrt{2} \left(\alpha \cdot C_{1\lambda}^{(0)}(\hat{r}) \right) / (cr^2)$ is the relevant relativistic coupling tensor [12].

After angular and many-body reduction, the scalar polarizability may be expressed as

$$\alpha_F^{(3)}(0) = \frac{1}{3} \sqrt{(2I)(2I+1)(2I+2)} \begin{Bmatrix} j_v & I & F \\ I & j_v & 1 \end{Bmatrix} \times g_I \mu_n (-1)^{F+I+j_v} (2T + C + R)$$

where g_I is the nuclear gyromagnetic ratio, μ_n is the nuclear magneton, $I = 7/2$ is the nuclear spin, and $j_v = 1/2$ is the total angular momentum of the ground state. The F -independent sums are ($|v\rangle \equiv |6s_{1/2}\rangle$)

$$\begin{aligned} T &= \sum_{m,n \neq v} \frac{(-1)^{j_m+j_v}}{2j_v+1} \frac{\langle v || D || m \rangle \langle m || D || n \rangle \langle n || \mathcal{T}^{(1)} || v \rangle}{(E_m - E_v)(E_n - E_v)} \delta_{j_n, j_v}, \\ C &= \sum_{m,n \neq v} (-1)^{j_m-j_n} \begin{Bmatrix} 1 & j_v & j_v \\ 1 & j_m & j_n \end{Bmatrix} \times \frac{\langle v || D || m \rangle \langle m || \mathcal{T}^{(1)} || n \rangle \langle n || D || v \rangle}{(E_m - E_v)(E_n - E_v)}, \\ R &= \frac{\langle v || \mathcal{T}^{(1)} || v \rangle}{2j_v+1} \left(\sum_{m \in \text{val}} - \sum_{m \in \text{core}} \right) \frac{|\langle v || D || m \rangle|^2}{(E_m - E_v)^2}. \end{aligned}$$

The summation indexes m and n range over valence bound and continuum many-body states and also over single-particle core orbitals. With this convention, the above expressions subsume contributions from intermediate valence and core-excited states and they also take into account so-called core-valence counter-terms [13]. Selection rules impose the following angular symmetries on the intermediate states: $s_{1/2}$ for $|n\rangle$, $p_{1/2,3/2}$ for $|m\rangle$ in the top diagram, $p_{1/2,3/2}$ for both $|m\rangle$ and $|n\rangle$ in the center diagram, and, finally, $p_{1/2,3/2}$ for the $|m\rangle$ in the residual term.

We will tabulate our results in terms of the conventional scalar Stark shift coefficient $k_s = -1/2 \left(\alpha_{F=4}^{(3)}(0) - \alpha_{F=3}^{(3)}(0) \right)$. For ^{133}Cs this coefficient

can be written more explicitly in terms of the F -independent diagrams as

$$k_s = -3 \left(\frac{2}{3} \right)^{5/2} g_I \mu_n (2T + C + R), \quad (2)$$

with the BBR coefficient $\beta = -4/15 (\alpha\pi)^3 T_0^4 / \nu_0 \times k_s$.

Numerical evaluation of the diagrams T, C , and R can be carried out either using the Dalgarno-Lewis method or by directly summing over individual intermediate states. Here we use the direct summation approach. This treatment is similar to the one used in high-accuracy calculations of atomic parity violation in ^{133}Cs [14]. The main advantage of this method is that one could explicitly exploit high-accuracy experimental data for energies, dipole-matrix elements, and hyperfine constants. When the accurate values are unknown, we use *ab initio* data of proper accuracy. In addition, this approach facilitates comparison with recent calculations [6, 7], which also use the direct summation approach.

The central technical issue arising in direct summation over a complete set of states is representation of the innumerable spectrum of atomic states. For example, even without the continuum, the bound spectrum contains an infinite number of states. A powerful numerical method for reducing the infinite summations/integrations to a finite number of contributions is the basis set technique. In particular, we employ the B-spline technique [15]. In this approach an atom is placed in a large spherical cavity and the single-particle Dirac-Hartree-Fock (DHF) orbitals are expanded in terms of a finite set of B-splines. The expansion coefficients are obtained by invoking variational Galerkin principle. The resulting set of the single-particle orbitals is numerically complete and finite. The technique has a high numerical accuracy and we refer the reader to a review [16] on numerous applications of B-splines for details.

We use B-spline set with 70 splines of order 7 for each partial wave and constrain the orbitals to a cavity of radius $R_{\text{cav}} = 220$ a.u. This particular choice of R_{cav} ensures that the lowest-energy atomic orbitals are not perturbed by the cavity. In particular, all core and valence DHF orbitals with radial quantum numbers 1 – 12 from the basis set produce energies and matrix elements in a close numerical agreement with the data from traditional finite-difference DHF code. These low-energy orbitals will produce true many-body states for a cavity-unconstrained atom.

To understand the relative role of various contributions, we start by computing the Stark shift at the DHF level. We obtain: $\frac{2T^{\text{DHF}}}{k_s^{\text{DHF}}} = 0.418$, $\frac{C^{\text{DHF}}}{k_s^{\text{DHF}}} = 0.003$, and $\frac{R^{\text{DHF}}}{k_s^{\text{DHF}}} = 0.518$, resulting in the Stark coefficient of $k_s^{\text{DHF}} = -2.799 \times 10^{-10} \text{ Hz}/(\text{V/m})^2$ [17]. It is clear that the top and residual terms dominate over the center diagram. The bulk (99.8%) of the value of the residual term is accumulated due to the principal $6s - 6p_{1/2,3/2}$ transitions. For the top term the saturation of the sum is

not as rapid, but still the dominant contributions come from the lowest-energy excitations: limiting the summations to the first four excited states recovers only 68% of the total value for the top diagram. In addition, we find that core-excited states contribute only 0.04% to the final value.

The above observations determine our strategy for more accurate calculations. We group the entire set of atomic states into the “main” low-lying-energy states (principal quantum numbers $n \leq 12$) and remaining “tail” states. We will account for the contribution from the “main” states using high-accuracy experimental and *ab initio* values. The contribution from the “tail” will be obtained using either DHF or mixed approach.

First, we describe the high-accuracy data used in our calculations. We need dipole and hyperfine matrix elements and energies. Experimental values for the dipole matrix elements for the following six transitions were taken from the literature (see compilations in Refs. [6, 18]) $6s_{1/2} - 6p_{1/2,3/2}$, $7s_{1/2} - 6p_{1/2,3/2}$, $7s_{1/2} - 7p_{1/2,3/2}$. Crucial to the accuracy of the present analysis were the matrix elements for the principal $6s_{1/2} - 6p_{1/2,3/2}$ transitions. We have used 0.005%-accurate value for $\langle 6s_{1/2} || D || 6p_{3/2} \rangle$ from Ref. [19]. The value for $\langle 6s_{1/2} || D || 6p_{1/2} \rangle$ was obtained by using the above $6s_{1/2} - 6p_{3/2}$ matrix element and 0.03%-accurate measured ratio [20] of these matrix elements. These six experimental matrix elements were supplemented by 92 values ($ns_{1/2} - n'p_{1/2,3/2}$ values for $n, n' = 6 - 12$) from high-accuracy *ab initio* calculations. We employ the relativistic linearized coupled-cluster singles-doubles (LCCSD) method. The underlying formalism, implementation, and results for alkali atoms are described in Ref. [18]. For dipole matrix elements the accuracy of the *ab initio* LCCSD method is a few 0.1%.

As to the high-accuracy values of the matrix elements of the hyperfine coupling, the diagonal matrix elements of the $\mathcal{T}^{(1)}$ tensor are directly related to the conventional hyperfine constants: $A = g_I \mu_n / j_v [(2j_v)/(2j_v + 1)/(2j_v + 2)]^{1/2} \langle v || \mathcal{T}^{(1)} || v \rangle$. We have used the compilation of hyperfine constants from Ref. [21] for the “main” $n = 6 - 12$ states. Off-diagonal matrix elements between the s -states were evaluated using the geometric-mean formula

$$\left| \langle ns_{1/2} || \mathcal{T}^{(1)} || n's_{1/2} \rangle \right| = \left\{ \langle ns_{1/2} || \mathcal{T}^{(1)} || ns_{1/2} \rangle \langle n's_{1/2} || \mathcal{T}^{(1)} || n's_{1/2} \rangle \right\}^{1/2}.$$

This expression has been shown to hold to about 10^{-3} in Ref. [22] (notice that the radiative corrections would start playing a role at a few 0.1% as well). If we had $6 \leq n \leq 12$ in the above expression, then the experimental value is used for its corresponding diagonal element on the right. If we also had $6 \leq n' \leq 12$, then that experimental value is also used for the corresponding diagonal element on the right, otherwise the DHF value is taken. (n and n' can be obviously interchanged in this prescription.) This

mixed approach has allowed us to uniformly improve the accuracy of the calculations. Indeed, in the numerically important top term, the hyperfine matrix elements come in the combination $\langle ns_{1/2} || \mathcal{T}^{(1)} || 6s_{1/2} \rangle$. As n grows, the correlations become less important, so the dominant correlation correction comes from the $6s_{1/2}$ state. Using the described mixed approach allows us to account for this dominant correlation. The geometric-mean formula holds only for the s states. For the off-diagonal matrix elements between various combinations of $6p_{1/2,3/2}$ and $np_{1/2,3/2}$ ($n = 6 - 9$) states we employed a modification of the LCCSD method augmented by perturbative treatment of the valence triple excitations (LCCSDpvT method) [18]. The accuracy of these matrix elements is a few %. As these matrix elements enter the relatively small center term, the effect on the overall theoretical error is negligible.

Finally, we used experimental energy values from the NIST tabulation [23], for states with principle quantum number $n = 6 - 12$, and the DHF values otherwise.

With the described set, we report our final result for the scalar Stark coefficient and relative blackbody radiation shift at 300K to be

$$\begin{aligned} k_s &= -(2.268 \pm 0.008) \times 10^{-10} \text{ Hz}/(\text{V/m})^2, \\ \beta &= -(1.708 \pm 0.006) \times 10^{-14}. \end{aligned} \quad (3)$$

The values of the individual diagrams are $\frac{2T}{k_s} = 0.449$, $\frac{C}{k_s} = -0.002$, and $\frac{R}{k_s} = 0.553$. When comparing with the DHF values, the most substantial modification due to correlations is in the center term, which changes the sign. Fortunately, this term is relatively small, and this extreme change does not substantially affect the final result.

The overall uncertainty of these results was determined from the uncertainties of the individual matrix elements and energy values used in their computation. Standard uncertainty analysis was done throughout all mathematical operations. For energy values taken from NIST, the uncertainty is assumed negligible. For all other experimental values, the reported uncertainty is used. For *ab initio* matrix elements (DHF, LCCSD, or LCCSDpvT) we assigned an assumed uncertainty. These assumed uncertainties were based on comparison between calculated and high-accuracy experimental values. This resulted in a relative uncertainty for both the scalar Stark coefficient and the BBR shift of 0.35%. We have performed several consistency checks, e.g., replacing experimental matrix elements and energies by *ab initio* LCCSD values or by replacing the DHF values for states with $n = 13 - 27$ with the LCCSD values. The final result was stable to such modifications within the stated uncertainty in Eq.(3). These tests provide us with additional confidence with respect to our standard error analysis based on errors of used experimental values. It is also worth noting that the present calculation does not include radiative corrections which may contribute at the level of a few 0.1% (some radiative corrections, e.g., vacuum polarization, are ab-

TABLE I: Values of k_S in $10^{-10}\text{Hz}/(\text{V/m})^2$.

		References
theory	-1.97 ± 0.09	Ref. [6]
theory	-2.06 ± 0.01	Ref. [7]
expt.	-2.05 ± 0.04	Ref. [4]
expt.	-2.271 ± 0.004	Ref. [3]
theory	-2.268 ± 0.008	present

sorbed in our final value already as we use *experimental* hyperfine constants).

A comparison with recent theoretical and experimental work is presented in Table I. While agreeing with the most accurate measurement by Simon et al. [3], our results are in substantial disagreement with the recent calculations [6, 7]. The principal differences between the present work and these calculations are: (i) more sophisticated treatment of correlations, and (ii) rigorous summation over the *complete* set of intermediate states in perturbative expressions. As discussed above, we used the numerically complete basis-set approach which approximates Rydberg states and continuum with quasi-spectrum. By contrast, in Ref. [6], the summations were truncated at $n = 9$, and in Ref. [7] at $n = 18$; neither work includes continuum. To illuminate the importance of the omitted contributions we truncate our summations

at $n = 12$. The resulting value deviates from our final k_s result by $0.29 \times 10^{-10} \text{ Hz}/(\text{V/m})^2$. This large 10% “continuum correction” brings the values from Refs. [6, 7] into essential agreement with our result. The fact that continuum needs to be included is hardly surprising, as, for example, about 20% of the textbook polarizability of the ground state of the hydrogen atom comes from the continuum states.

To conclude, here we have reported results of relativistic many-body calculations of the BBR shift, one of the leading systematic correction in ^{133}Cs frequency standard and a subject of the recent controversy. Our 0.35%-accurate result re-validates high-precision Stark shift measurements [3]. Our work also clarifies the origin of the reported discrepancy between that measurement and recent calculations [6, 7].

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Note. While completing writing this manuscript, we have learned of another accurate many-body calculation of the BBR shift in ^{133}Cs clock [24]. Their result, $k_s = 2.26 \times 10^{-10} \pm 1\% \text{ Hz}/(\text{V/m})^2$, is in agreement with our more accurate value.

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